

Effect of Pressure Washing Conditions on the Removal of Cs from Urban Surfaces

ASSESSMENT AND EVALUATION REPORT



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Assessment and Evaluation Report

National Homeland Security Research Center
Office of Research and Development
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

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List of Acronyms and Abbreviations

ASTM	American Society for Testing and Materials, now ASTM International
ANCOVA	Analysis of Covariance (Model)
ANOVA	Analysis of Variance (Model)
Ba	Barium
Ce	Cerium
CeO	Cerium Oxide
Co	Cobalt
COTS	Commercial Off-The-Shelf
Cs	Cesium
CsCl	Cesium Chloride
DI	Deionized
EPA	U.S. Environmental Protection Agency
gpm	Gallon(s) per Minute
GLM	General Linear Model
HNO ₃	Nitric Acid
HSRP	Homeland Security Research Program
ICP-MS	Inductively Coupled Plasma - Mass Spectrometry (meter)
ID	Identification
ITSD	Internal Standard
LIMS	Laboratory Information Management System
MB	Method Blank
Mg	Magnesium
NHSRC	National Homeland Security Research Center
Pb	lead
PDS	Post Digestion Spike
psi	Pound(s) per square inch

QA	Quality Assurance
QAPP	Quality Assurance Project Plan
QC	Quality Control
QCS	Quality Control Standard
RDD	Radiological Dispersal Device
RH	Relative Humidity
Rh	Rhodium
RSD	Relative Standard Deviation
Sr	Strontium

Executive Summary

The U.S. Environmental Protection Agency's (EPA) Homeland Security Research Program (HSRP) helps protect human health and the environment from adverse impacts of terrorist acts by carrying out performance tests on homeland security technologies. This study investigated the impact of water pressure conditions for decontamination of urban surfaces contaminated with cesium (Cs). The contaminated surfaces were prepared with two different deposition methods to control the penetration depth of Cs using water and methanol as solvents; the surfaces were then washed in a chamber, simulating the delivery of high pressure water. Various water pressures, applied in both vertical and horizontal orientations of the surface, were evaluated for efficacy against the contaminated coupon surfaces. The amount of Cs in the liquid and solid portions of the water rinsate samples was measured and analyzed to determine the impact of individual wash conditions.

The main goal of this project was to determine the impact of water pressure conditions in the decontamination of urban surfaces contaminated with Cs particles. Various conditions of water pressure, deposition type, and surface orientation were tested to investigate the impact on Cs removal from three different urban surfaces. Increased water pressure improved Cs removal efficacy for all three surfaces. Total removal efficacy varied by surface type and deposition method. Asphalt showed the highest removal efficacy for Cs (42 % to 78 %), followed by brick (5 % to 35 %), and concrete (6 % to 25 %). Increased Cs removal was observed from dry (methanol) deposition as opposed to wet (water) deposition for all three surfaces. This increased Cs removal may be related to the penetration depth of Cs after deposition. Methanol evaporates more quickly than water, and as a result, the majority of the Cs remains close to the surface and becomes easier to remove. Orientation was not a significant factor for either liquid or solid removal for all three surfaces. The Cs removal mechanism was further investigated by separate measurement of the amount removed in solid phase and liquid phase.

The results showed that when comparing Cs amount in solid samples for all three substrates for the same conditions, asphalt showed decreasing Cs amount with increasing pressure, but brick and concrete showed increasing Cs amount with increasing pressure. Comparison of the Cs amount in liquid samples for the same conditions showed the same profound effect for asphalt as total efficacy, whereas brick and concrete showed a minimal increase in Cs amount with increasing pressure.

The dry and wet deposition test results demonstrated that delayed pressure washing may lead to decreased removal of Cs from the surface due to the subsurface penetration. For asphalt, Cs removal in the liquid phase was dominant, and the removal efficacy increased with increased water pressure up to 7000 psi. The water pressure (6000 to 7000 psi) provided high decontamination efficacy (50 % to 80 %) with minimal surface degradation. In the case of brick and concrete, test results showed a minimal increase (less than 10 % increase) in Cs removal as a function of pressure in the range of 4000 to 7000 psi. However, analysis of the data showed that increased removal efficacy for brick and concrete were related to removal of solid materials by the high pressure stream. Extensive layer removal from brick and concrete surfaces is expected to increase the removal efficacy because of subsurface penetration and Cs sorption on the removed surfaces.

1 Introduction

A radiological dispersal device (RDD), also called a dirty bomb, is the combination of a conventional explosive device with radioactive materials that can be obtained from industrial, commercial, medical, or research applications.^{1,2} An RDD attack can impact a society in various ways including creation of casualties, disruption of the economy, and relocation of people from the contaminated area.^{3,4,5} Fast and cost-effective decontamination strategies are critical to minimize the social and economic damage from an RDD event, increasing the community resilience.

The U.S. Environmental Protection Agency (EPA) has conducted a series of tests to investigate the fate and transport of RDD materials on urban surfaces under varied conditions.^{6,7,8,9,10} The results from these previous studies showed varied fates for cesium (Cs) on surfaces, depending on contaminant deposition conditions, surface types, and environmental conditions. A follow-up study¹¹ focused on the assessment of various conditions of water wash down parameters, including wash down duration, water pressure, wash angle, and wash patterns on asphalt, brick, and concrete using cesium chloride (¹³³CsCl) particles. The results showed a positive correlation with wash duration and water pressure for the asphalt and concrete samples and demonstrated that a 90-degree wash angle was more effective than a 45-degree wash angle for the brick and concrete samples. However, these findings are applicable primarily to the conditions where Cs particles have limited penetration into porous surfaces.

The current study tested water pressure in an effort to optimize the pressure washing methods to increase the removal efficacy on these same three urban surface materials (asphalt, brick, and concrete). The research focused on the assessment of various water pressures for Cs removal as functions of deposition methods, surface types, and water application orientations.

2 Materials and Methods

2.1 Test Overview

The amounts of Cs removed from urban surfaces via high pressure washing were studied under various application conditions. Various water pressures, Cs deposition methods, and vertical and horizontal surface orientations were explored. Three different building materials were contaminated with Cs particles via aerosolization, and the contaminated surfaces were washed using a high pressure washer. The liquid and solid (filtered) portions of the rinsate samples were analyzed for Cs amounts that were removed from the surface.

2.2 Building Materials

Building materials are described in Table 2-1. Concrete coupons (12" x 12" x 1.5" [W x L x H]) were prepared according to the method used in the previous Water Wash Down study.¹¹ Each coupon was visually inspected and any coupons with defects, cracks, or stains were discarded. Concrete coupons were prepared following the manufacture instructions (Quikrete® Portland Cement Atlanta, GA). Brick coupons (12" x 12" x 2.25" [W x L x H]) were prepared by a brick mason. The brick walls were built using mortar and with the same method employed when constructing a home. Asphalt coupons (6" diameter) were supplied by the North Carolina Department of Transportation. Since the asphalt coupons were from actual roadways, thickness varied, but test coupons were chosen to be as close to a height of 1.5 inches as possible. Figure 2-1 shows the finished concrete, brick, and asphalt coupons.

Table 2-1. Building Material Descriptions and Sources

Material	Description	Locality	Source
Brick	Red, fine-grained	Made from North Carolina red Triassic clay	Triangle Brick Company, Durham, North Carolina
Concrete	Sand, Portland cement mix	N/A	Home Depot, North Carolina
Asphalt	Laboratory pressed asphalt	N/A	North Carolina Department of Transportation



Figure 2-1. Finished concrete (a), brick (b), and asphalt (c) coupons

Five sides of each concrete and brick coupon and the edges and back of the asphalt coupons were sealed with water-impermeable sealant (Stonelok™ E3, Richard James Specialty Chemicals Corp., Hastings on Hudson, NY). The top surface remained unsealed for deposition of surrogate RDD contaminants and application of pressurized water. The sample identification (ID) was marked on two opposite sides and the back of each coupon with paint markers before sealing.

2.3 Coupon Conditioning

Concrete coupons were poured and cured under ambient conditions for at least 28 days before being prepared for surface contamination as described below. Prior to particle deposition, the top surface of all coupons was cleaned thoroughly with a 2300 psi/2.3 gallons per minute (gpm) pressure washer to remove any loose pieces of building material. After cleaning, the coupons were dried in laboratory air for one week. Daily moisture measurements were then taken for one week after the coupons were allowed to dry in the laboratory (Dawson Multi-Function Moisture Meter, Model DSM170, Diamond Bar, CA). Minimal differences in moisture were observed and appeared to be a function of room relative humidity (RH). The moisture measurements are discussed in Section 3.2. After deposition, coupons were stored in laboratory air (23 ± 2 °C and 40 ± 2 % RH) for 24 hours prior to use in the pressure washer testing.

2.4 Cesium Particle Deposition

A solution containing Cs particles in deionized (DI) water (wet) or methanol (dry) was deposited onto coupons using a metered syringe (MicroSprayer® Aerosolizer, Model 1A-1C and FMJ-250 High Pressure Syringe, Penn Century, PA). Dry Cs deposition mimics the surface contamination status immediately after contamination and wet Cs deposition represents the surface contamination condition after rain or high relative humidity. It is expected that dry deposition causes less Cs subsurface penetration than wet deposition due to fast evaporation rate of methanol compared to water. Templates were used to ensure that the Cs was deposited over the majority of the surface of the coupon in a fixed-array pattern. The tip of the high pressure syringe was positioned 5 inches from all coupon surfaces during deposition. To determine the optimum deposition height, multiple tests were conducted using food coloring in water and methanol. The colored solution was deposited on white paper at various heights. It was desired to have the deposition pattern be well dispersed, but not producing a spray pattern that impacted the sides of the deposition template, nor producing a semi-direct deposition pattern onto the white paper. Figure 2-2 shows the nine-hole template used for the 12-inch by 12-inch concrete and brick coupons. The openings are pinhole sized, but the shape of the grid can be inferred from the location of the high pressure syringe tips placed on the diagonal. Figure 2-3 shows the two-hole template used for the six-inch diameter asphalt coupons. In all cases, each template hole was dosed with 25 μ L of the 200 ppm CsCl solution (wet or dry) and allowed to dry under ambient conditions for 24 hours before testing. This procedure yields a nominal deposition of 45 μ g of CsCl onto the concrete and brick coupons and 10 μ g of CsCl onto the asphalt coupons. The deposition amount was calibrated (spike control) by depositing the cesium solutions onto a 5 x 5 array of clean polyethylene plastic sheets with the syringe tip at the same distance as the building material coupons. Each set of spike control samples for Cs was transferred to a clean 1000-mL polyethylene beaker for extraction. The beaker was filled with 1 % ultrapure OPTIMA nitric acid (HNO₃) (Sigma-Aldrich®, St. Louis, MO) in DI water until the solution covered the plastic surface entirely (~360 mL). The spike control samples were extracted by sonication for 20 minutes. After sonication, the plastic squares were removed and discarded. The beaker was filled to 450 mL with HNO₃. Using an Eppendorf pipette, 0.5 mL was extracted from the beaker and placed into a clean 50-mL vial. The vial was then filled to a volume of 50 mL with DI water and submitted for analysis by Inductively Coupled Plasma - Mass Spectrometry (ICP-MS).

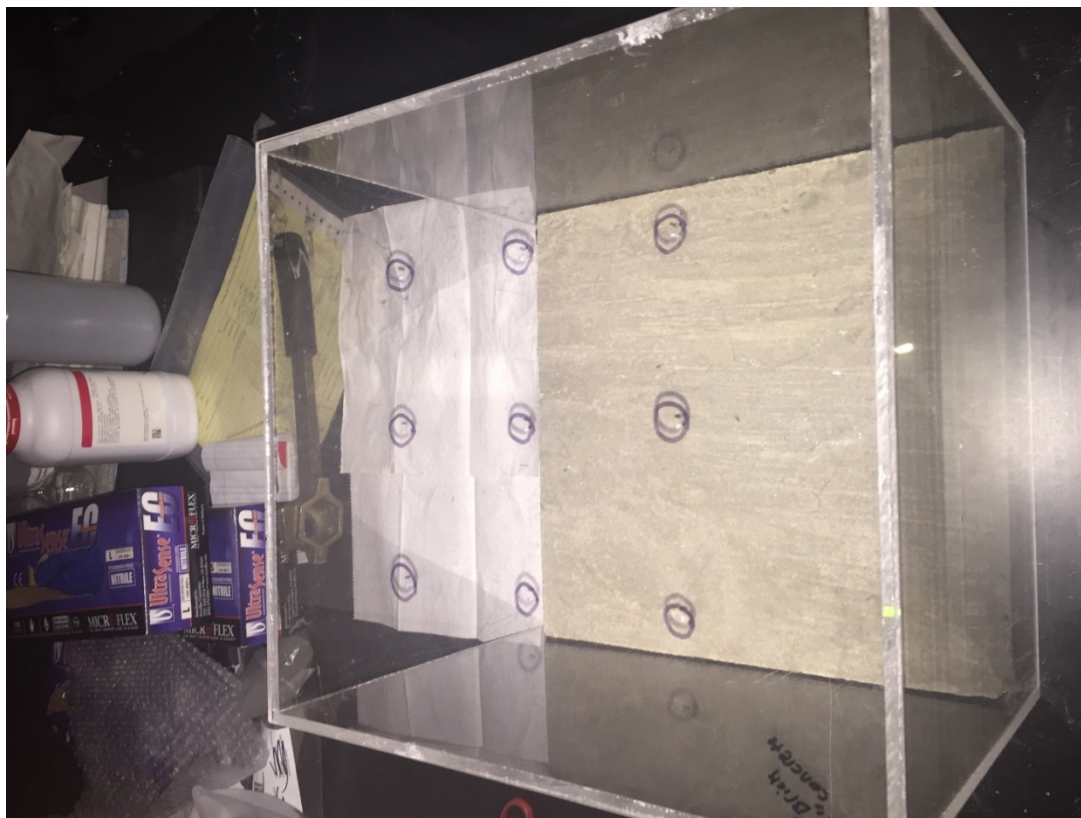


Figure 2-2. Particle deposition onto a brick and concrete coupon



Figure 2-3. Particle deposition onto an asphalt coupon

2.5 Pressure Washing System

The contaminated coupons were subjected to pressure washing using a high-pressure washer system (Silver Eagle Manufacturing, PO Box 7346, Cut-N-Shoot, Texas, 77306; Engine Driven High Pressure Washer System) within 24 hours after contamination. Triplicate coupons were prepared for each pressure-washing test condition for brick and concrete. Six coupons were prepared for each pressure-washing test condition for asphalt. After a test, coupons were allowed to dry in laboratory air.

All coupons were washed in a chamber using a high-pressure water-delivery system (Figure 2-4) and 25-degree fan angle for each nozzle. A stainless steel chamber (4' x 4' x 4') was used and is shown in Figure 2-5. The nozzle was fixed in the center of a chamber side and chamber top to accommodate both vertical and horizontal coupon holders that were positioned on the bottom or back wall of the chamber. In Figure 2-5, a brick coupon can be seen in the horizontal position; the coupon holder for the vertical orientation coupons can be seen on the back wall. Figure 2-6 shows an asphalt coupon in its holder. The front door of the chamber was opened to allow cleaning of the inside of the chamber after each test. The chamber bottom was slanted on all four sides to collect the rinsate water. To ensure accurate and consistent delivery of pressure during testing, the chamber was fitted with two pressure plates, one each in the horizontal and the vertical orientation. At the beginning and end of each day of testing, the pressure measurement for each nozzle and water pressure being tested that day.

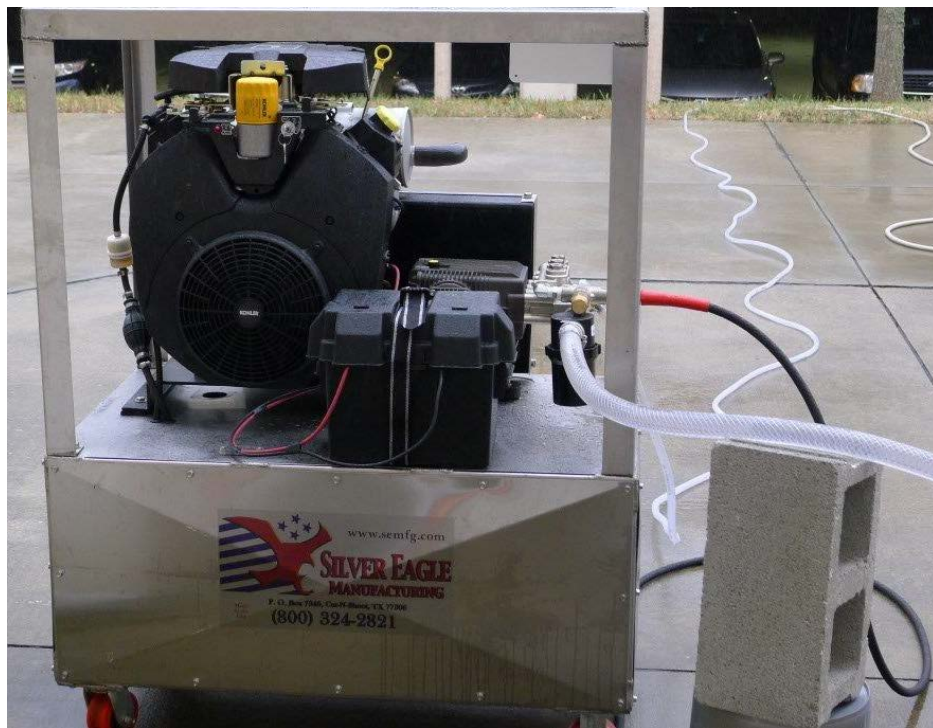


Figure 2-4. High-pressure washer system

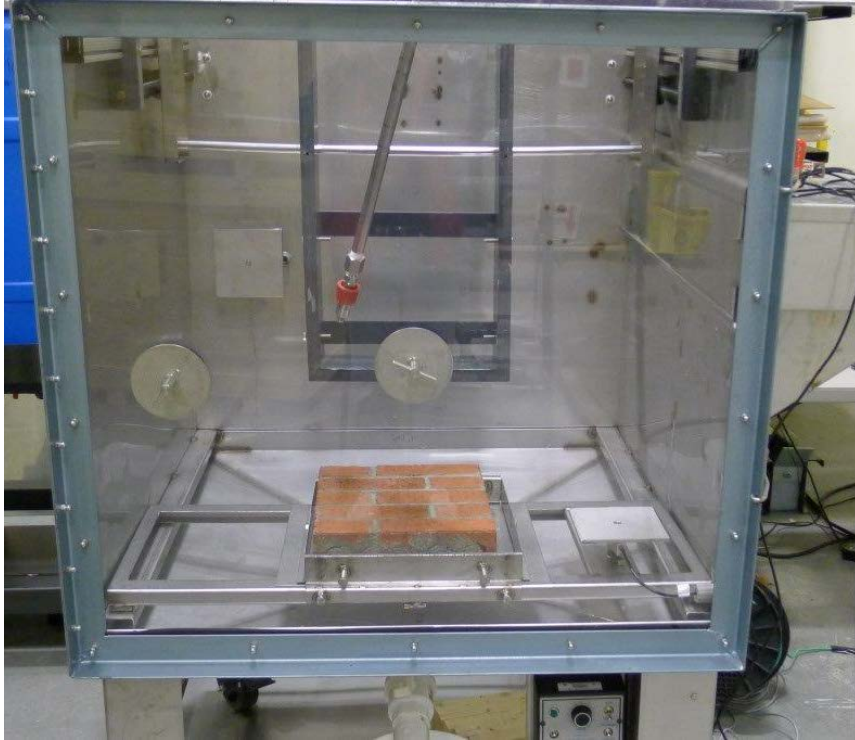


Figure 2-5. Pressure washing chamber

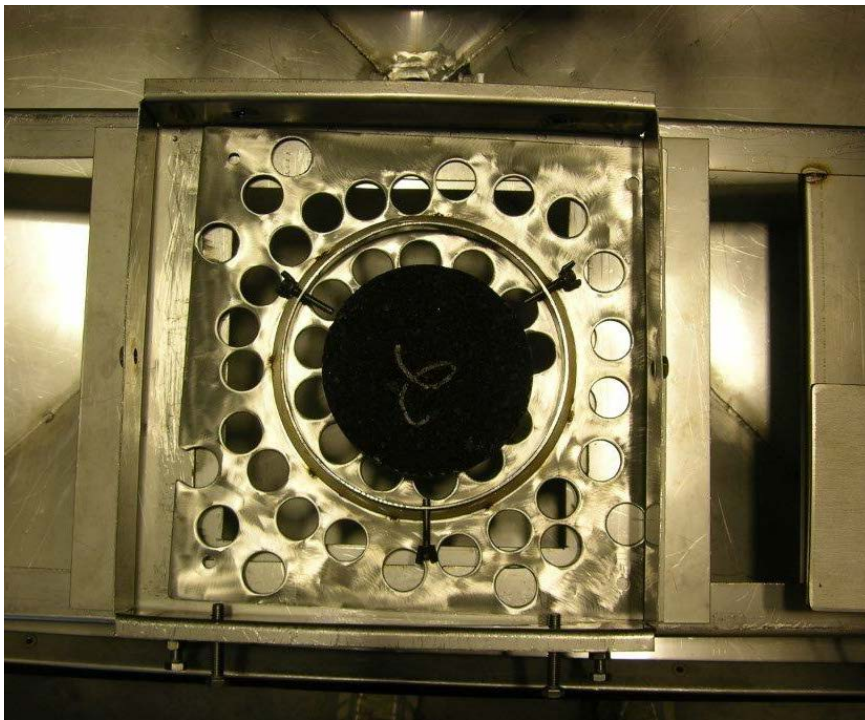


Figure 2-6. Asphalt coupon in the chamber holder

DI water was used for the pressure-washer tests, and the water was applied for 10 seconds at a distance of ten inches from the coupon surface. The rinsate water was collected in clean plastic containers (Cubitainer®, Fisher Scientific, Pittsburgh, PA) located under the bottom drain. The rinsate containers were weighed to record the amount of water collected during each pressure-washer test; the containers were then capped and submitted to the laboratory for filtration and analysis of both the solid and liquid portions by ICP-MS. After the test, the coupons were removed from the chamber coupon holder, and the chamber surfaces were pressure-washed with a ten-second DI water rinse. This blank chamber rinse was also collected, and a 50-mL vial was submitted to the laboratory for Cs analysis. As an additional precaution against cross-contamination, the inside chamber was further rinsed with DI water using a garden hose for 25 to 30 seconds, taking care to clean out any particulate matter remaining around the drain. This final rinse was collected in a five-gallon bucket and discarded.

2.6 Wash Down Conditions

2.6.1 Pressure Washing

All coupons were washed for a duration of 10 ± 1 seconds and at a 90-degree angle between water jet direction and coupon surface. Coupons were tested in both vertical and horizontal orientations for brick and concrete; asphalt was tested only in the horizontal orientation. Figure 2-7 shows a brick coupon being sprayed from the top of the chamber. Other test conditions were deposition method (wet or dry) and the water pressure (psi) being tested. Water from the pressure washer covered the entire coupon surface. The rinsate volume for the ten-second washes was collected into a single one-gallon Cubitainer. The rinsate samples were analyzed for amount of Cs to determine whether the decontamination efficacy is affected by the coupon orientation at various water pressures, using both wet and dry deposition methods and coupon orientation.



Figure 2-7. Horizontal coupon being sprayed in chamber

2.6.2 Water Pressure

Five different water pressures were applied to the coupons from the same distance and duration. The applied nozzle water pressures were 2100, 4000, 5000, 6000, and 7000 psi. Pressure was adjusted by changing the nozzle type. The 2100 psi/2.2 gpm pressure was achieved by using a Commercial Off-The-Shelf (COTS) pressure washer purchased from Lowes (Troy-Bilt, Briggs and Stratton Power Products, PO Box 702, Milwaukee, WI). The high pressure washer used high pressure ceramic nozzles, and the flow ranged from 4.5 to 7.9 gpm. The rinsates from varied water pressure tests were analyzed for Cs amount to determine the dependence of decontamination efficacy on water pressure.

2.6.3 Wash Pattern

A single wash pattern was tested on all coupons. Figure 2-8 shows the pattern proceeding from top to bottom. The pressure wash duration was ten seconds, wash angle was 90 degrees, and distance from the nozzle tip to the coupon surface was ten inches.

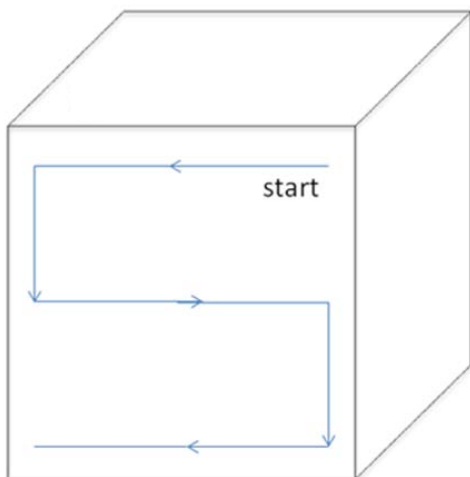


Figure 2-8. Water wash down test pattern

2.7 Test Matrix

The coupon test matrix is shown in Table 2-2. Three samples of each building material were prepared for both deposition methods and coupon orientation, with the exception of asphalt. Since asphalt is encountered in real-life use only in the horizontal orientation, the vertical orientation was not tested for this material. As shown in Table 2-2, a total of three blank coupons were prepared for each test scenario, except six coupons were prepared for asphalt. Blank coupons were coupons without Cs deposition. These blank coupons were washed down and the rinsate samples were collected for baseline Cs solid and liquid concentration determination.

Table 2-2. Test Matrix for Pressure Washing Tests

Material	Deposition Condition	Orientation	Building Material Samples
Fine Aggregate Concrete	Wet	Horizontal	3
	Dry	Horizontal	3
	Wet	Vertical	3
	Dry	Vertical	3
	Blank	Horizontal	3
	Blank	Vertical	3
Brick Wall	Wet	Horizontal	3
	Dry	Horizontal	3
	Wet	Vertical	3
	Dry	Vertical	3
	Blank	Horizontal	3
	Blank	Vertical	3
Asphalt	Wet	Horizontal	6
	Dry	Horizontal	6
	Blank	Horizontal	6

2.8 Analysis of Pressure Wash Rinsates

Standard methods modified by the laboratory for this work included EPA Method 6020A, “Inductively Coupled Plasma-Mass Spectrometry” and EPA Method 3050B “Acid Digestion of Sediments, Sludges and Soils”. The custom methodology consisting of the analysis of rinse water samples via ICP-MS was developed by Pace Analytical Services, Inc. (6701 Conference Drive, Raleigh, NC 27607) and implemented by First Analytical Laboratories (7517-101 Precision Drive, Raleigh, NC 27617) to determine and quantify the presence of ultra-trace elemental content of Cs, cobalt (Co), and strontium (Sr). A custom methodology consisting of the analysis of particulate matter obtained from the filtration of rinse water samples via ICP-MS was developed and implemented to determine and quantify the presence of ultra-trace elemental content, specifically Cs. Results were reported for use in the determination of rinse procedure efficacy when carried out utilizing various rinsing techniques on an assortment of substrate types.

The test rinsate samples, in one-gallon high-density polyethylene Cubitainers, were delivered in their entirety to the analytical laboratory (First Analytical Labs, Raleigh, NC). For the blank chamber rinsate samples, a 50-mL polyethylene centrifuge tube was submitted, and the remainder of the blank rinsate sample was discarded. The samples were subjected to a visual validity assessment by the laboratory to ensure that no samples were compromised during transit (i.e., leaked sample, loose sample container lid, cracked sample container, etc.). Following the validity check, the samples were compared to the total number of samples denoted on the chain-of-custody form to ensure that all samples were present at time of receipt.

First Analytical Labs analyzed both the rinsate and filtered (solid) portions of each sample using a custom methodology developed by Pace Analytical Services (Raleigh, NC) for the ultra-trace analysis of Cs by ICP-MS: "ICP-MS Analysis of Filtered Rinse Water for Ultra-Trace Levels of Cesium (Cs), Cobalt (Co) and Strontium (Sr), RAL-0001-MET, Revision 1" and "ICP-MS Analysis of Particulate Matter Obtained From Filtered Rinse Water for Ultra-Trace Levels of Cesium (Cs), RAL-0002-MET, Revision-New". These methods were modified versions of EPA Standard Methods 200.8 and 6020A and are summarized in more detail below. The modified methods were verified by the laboratory before use. A model ELAN DRC-e ICP-MS (Perkin Elmer, Concord, Ontario) was used for Cs analysis. The typical operating conditions of ICP-MS are summarized in Table 2-3.

Table 2-3. Typical Operating Conditions for ELAN DRC-e

Parameter	Operating Condition
Pulse Stage Voltage	1700 Volts
Analog Stage Voltage	-2100 Volts
Radio Frequency Generator Power	1200 Watts
Lens Voltage	7 Volts
Vacuum Pressure	4.5 e -005 Torr
Nebulizer Gas Flow Rate	0.96 L/min
Auxiliary Gas Flow Rate	15.0 L/min
Nebulizer Type	Ryton Scott Cross Flow Spray Chamber with Alumina Injector Tube
Ambient Temperature	72.0 °F ± 5.0 °F (22.2 °C ± 2.8 °C)
Sampler and Skimmer Cone Type	Platinum
Mass Spectrometer Type	Quadrupole
Detector Type	Dual Mode Electron Multiplier

2.8.1 Rinsate Sample Preparation

Following the custody sign-over process, the samples were logged into the First Analytical laboratory information management system (LIMS). Each rinsate sample went through seven main process phases once received and logged into the LIMS: (1) sample preparation, (2) sample analysis, (3) data interpretation, (4) data entry, (5) preliminary data submission, (6) data validation, and (7) final report generation/distribution.

2.8.2 Solid Sample Preparation

Each solid sample went through nine main process phases once received and logged into the LIMS: (1) sample filtration, (2) sample digestion, (3) sample digest preparation, (4) sample analysis, (5) data interpretation, (6) data entry, (7) preliminary data submission, (8) data validation, and (9) final report generation/distribution.

Prior to beginning the analytical process, the samples were filtered using a standard manual vacuum system to separate the solid portion of the sample from the aqueous portion of the sample. The liquid

obtained from the filtration process was retained and stored in high-density polyethylene sample containers for subsequent analysis as described in Section 2.8.3. The solid material obtained from the filtration process was collected on acid-washed filter papers and placed in a laboratory oven at 100 ± 10 °C to dry for a minimum of four hours.

Upon completion of the drying process, the filters with the retained solids were removed from the oven and allowed to cool to ambient temperature under a clean fume hood. Dried and cooled filters were ground into a consistency that was more easily digested and weighed out into pre-labeled digestion vials using an analytical balance. In addition to the samples, there were two digestion vials labeled “Method Blank (MB)” and “Post Digestion Spike (PDS)”. The vial labeled MB was prepared by weighing out 10 grams of ASTM International (ASTM) Type I water, and the vial labeled PDS was prepared by cutting up one raw filter and using an analytical balance to determine the weight of the raw filter added to the vial. All sample weights were recorded on each digestion vial as well as on the applicable sample preparation bench sheet.

All digestion vials were then placed into a polycarbonate digestion rack, covered with a watch lid, and transported to a fume hood. While under the fume hood, 10 mL of a 1:1 ultra-trace metal grade nitric acid/ASTM Type I water solution was added using an adjustable 1-to-10-mL pipette, re-covered with watch lids, and placed into a block digestion system (Environmental Express, Charleston, SC, Model SC-100) pre-heated to 105 ± 1 °C. The samples were heated for approximately 30 minutes, removed from the block digestion system, and allowed to cool to room temperature under a closed sash fume hood. Once cooled, 5 mL of ~70-percent ultra-trace metal grade nitric acid (HNO_3) was added to each vial, and the rack of covered samples was returned to the block digestion system and pre-heated to a temperature of 95 ± 1 °C. The samples were refluxed until all brown fumes subsided, adding additional 5-mL aliquots of HNO_3 as necessary. Once all brown fumes were absent, the samples were removed from the block digestion system and allowed to cool to room temperature, after which 2 mL of ASTM Type I water and 2 mL of hydrogen peroxide (H_2O_2) were added to each vial, respectively, and the covered vials were returned to the block digestion system and allowed to heat for 30 minutes at a temperature of 95 ± 1 °C. After a period of 30 minutes, the samples were visually checked for signs of effervescence. Should such signs be observed, additional water and H_2O_2 were added to the vials in 2-mL aliquots, and the samples were allowed to heat for additional 30 minute intervals. This process was continued until no signs of effervescence remained.

Upon completion of the effervescent phase, the samples were heated on a hot plate for a period of 60 minutes, making sure that samples did not reduce to dryness. The samples were cooled to ambient conditions under a closed sash fume hood and then filtered using clean funnels lined with quantitative-grade, ashless filter paper and washed with ASTM Type I water. The filtrate was collected into a clean labeled vial and brought to a final volume of 50 mL using ASTM Type I water. Prior to capping the samples, the PDS vial was spiked with 50 μL of the secondary source Cs certified reference material. The samples were then transferred for analysis via ICP-MS as described in Section 2.8.3.

2.8.3 Sample Analysis

Following the completion of a successful calibration and quality control (QC) check standard routine (see Section 3.1), each batch of samples and controls was loaded onto the ICP-MS for analysis. The samples were prepared by pipetting 10 mL of a well-mixed sample, as well as 10 μL of an internal standard solution, and then dispensing both liquids into a pre-labeled polyethylene test tube. After all samples

within a given batch were prepared, all test tubes were capped and shaken thoroughly to ensure even dispersion of the internal standard solution within the sample aliquot. In addition to the preparation of batched samples, the appropriate number of control and duplicate samples were prepared and included in the batch prior to loading the samples onto the instrument for analysis.

Any sample that yielded a result greater than the highest standard on the calibration curve was re-analyzed using the lowest dilution factor necessary to bring the result to a concentration within the calibration curve range. Any sample that yielded a result significantly impacted by matrix interferences was repeated using the lowest dilution factor necessary to reduce or eliminate the interfering component(s) to a level where the potential for erroneous signal amplification or suppression was mitigated. All internal standard recovery values were monitored to ensure that there were no sample introduction issues, instrument drift, or other problems that have the potential to effect data quality.

2.8.4 Laboratory Data Entry, Validation, and Reporting

All sample and QC data were entered into a customized Excel-based spreadsheet. All sample- and batch-related QC data were included.

2.8.5 Laboratory Calculations

All internal standard (ITSD) recovery calculations used the following formula:

$$[\text{ITSD (Sample)}/\text{ITSD (Calibration Blank)}] * 100$$

All quality control standard (QCS) isotopic recovery calculations used the following formula:

$$[\text{QCS}_{\text{Obtained Conc.}}/\text{QCS}_{\text{Target Conc.}}] * 100$$

All sample/duplicate relative percent difference calculations used the following formula:

$$[(\text{Result}_1 - \text{Result}_2)/\text{Result}_{\text{Avg}}] * 100$$

All solid sample results were calculated using the following formula:

$$\text{Result } (\mu\text{g/Kg}) = [\text{Result (mg/L)} * 50 / \text{Sample Aliquot (g)}] * 1000$$

2.9 Cs Removal Efficacy

The efficacy of a pressure washer test was assessed by determining the amount of Cs in the water wash rinsate samples. The Cs amount in the rinsate water samples was compared to the Cs amount in the positive control rinsate. Removal efficacy of Cs from the coupon material was calculated as the ratio of M_r and M_{PC} :

$$\text{Total Cs Removal Efficacy (\%)} = (M_{SR} + M_{LR}) / M_{PC} \times 100$$

where M_{SR} and M_{LR} are the average Cs amounts (μg) in solid and liquid phases in triplicate rinsate samples, respectively, and M_{PC} is the average Cs amount (μg) from five positive controls.

2.10 Statistical Methods

Statistical analyses were performed on the data set to determine statistical significance of results. The specific tests performed are discussed below.

2.10.1 *P-value*

The p-value is defined as the probability of obtaining a result equal to or more extreme than the value actually observed. The p-values calculated for these analyses were set at the 95 % confidence level. Therefore, if an obtained p-value is lower than 0.05, the set of data being examined is significant at the 95 % confidence level.

2.10.2 *ANCOVA Analysis*

A general linear model (GLM) (Snedecor and Cochran, 1980, Statistical Methods¹²) was used to evaluate the Cs removal testing. The following categorical independent variables were evaluated as factors in the model: orientation (H, V), material (asphalt, brick, concrete), and deposition (wet, dry). These treatment factors represent the categorical independent variables in the model. The testing was carried out at five different pressure levels, and pressure was evaluated as a continuous independent variable. The dependent variables evaluated were liquid Cs removed, solid Cs removed, and total Cs removed. This modeling approach is commonly referred to as an analysis of covariance model (ANCOVA). ANCOVA models combine the features of analysis of variance models (ANOVA) and linear regression models and are used to evaluate treatment differences when there are both categorical and continuous treatment factors in an experiment (Snedecor and Cochran, 1980). Both main and crossed effects were evaluated for statistical significance. A post-hoc multiple-comparison test (Tukey [1953])¹³ was used to evaluate the model-predicted means for each factor for both main and crossed effects when significant. All modeling was conducted using the GLM procedure of SAS 9.3 statistical software (SAS Institute, Cary, NC, 2012¹⁴).

3 Quality Assurance/Quality Control

Quality assurance/quality control (QA/QC) procedures were performed according to the EPA-approved quality assurance project plan (QAPP) for this test. All equipment and monitoring devices used at the time of evaluation were verified as being calibrated.

3.1 Calibration

3.1.1 Pressure Plate Calibration

The wash chamber was equipped with two pressure plates, one mounted horizontally and one mounted vertically. The pressure plates were attached to a computer program that logged the actual pressure in pounds being exerted on the plate by the pressure washer nozzle for a given pressure. During each day of testing, multiple tests of pressure were conducted by spraying onto the pressure plate for approximately 20 seconds. In addition, a 25-pound kettle bell was placed on the horizontal plate to provide a consistent measurement for each calibration test. The pressure plate calibration was performed before and after each day of testing to document any drift in the pressure reading during the actual testing. Each pressure plate calibration test at each pressure consisted of a minimum of 10 individual measurements. The data collected were used to calculate an average at each pressure level for testing and a relative standard deviation (RSD). The results of these pressure tests, for both horizontal and vertical orientations, are presented in Tables 3-1 and 3-2, respectively.

Table 3-1. Horizontal Pressure Plate Calibration Tests

	Before		After	
Measurement (lb)	Average (lb)	RSD (%)	Average (lb)	RSD (%)
25	25.02	0.4	24.98	0.4
2100	4.15	12.7	4.19	9.2
4000	20.86	16.7	22.21	8.4
5000	21.61	6.2	22.50	5.2
6000	23.17	9.2	23.47	5.5
7000	22.95	7.3	23.49	5.0

Table 3-2. Vertical Pressure Plate Calibration Tests

	Before		After	
Measurement (lb)	Average (lb)	RSD (%)	Average (lb)	RSD (%)
2100	3.51	25.9	3.00	22.7
4000	22.53	4.5	21.62	9.7
5000	21.88	8.6	19.45	12.8
6000	23.10	10.7	23.26	6.1
7000	22.79	10.0	22.61	7.5

3.1.2 ICP-MS Calibration

A daily performance assessment was carried out on the ICP-MS prior to the analysis of any samples. This assessment was performed using a multi-element certified reference material (Perkin Elmer Smart Tune solution, 10.0 µg/L of various elements – Perkin Elmer Catalog #N8125040) obtained from the instrument manufacturer. Upon completion of a valid daily performance assessment, the instrument was calibrated for the element of interest using one of the following methods: Low Level Cesium Method, Low Level Cobalt Method, or Low Level Strontium Method.

All of the above methods employed a four-point, external, linear-through zero calibration type with curve points of 0.025, 0.50, 5.0 and 10.0 µg/L; each method utilized an isotope (other than the element of interest) to serve as an internal standard.

Prior to instrument calibration and data analysis, instrument performance was validated by performing a daily performance assessment. The criteria for a passing daily performance assessment included:

- Rhodium (Rh) counts per second must be $\geq 150,000$
- Lead (Pb) counts per second must be $\geq 100,000$
- Magnesium (Mg) counts per second must be $\geq 20,000$
- Cerium (Ce)/Cerium Oxide (CeO) ratio should be $\leq 3.00 \%$
- Barium (Ba)⁺⁺ should be $\leq 5.00 \%$
- Combined 220 mass background must be < 30 counts per second
- RSD among all replicate sweeps for each tune component (sans 220 background) must be $< 5.00 \%$

All calibration curve correlation coefficient numbers were ≥ 0.9995 . The calibration curve was verified at the beginning and at the end of the analytical sequence by analyzing a blank QC standard (0.000 µg/L) that yielded a result < 0.025 µg/L, a low-range QC standard (0.500 µg/L) that yielded a result between 0.450 and 0.550 µg/L, a mid-range QC standard (5.00 µg/L) that yielded a result between 4.50 and 5.50 µg/L, and a high-range QC standard (10.0 µg/L) that yielded a result between 9.00 and 10.0 µg/L. The percent recovery for all applicable internal standards must be between 70 % and 125 %. Sample batch sizes did not exceed 20 samples, and each batch of 20 samples was bracketed by one blank QC and one low-range, mid-range, or high-range QC standard, making sure to rotate between all three range standards with each additional batch analyzed. In addition to batch-associated QC standards, one sample duplicate was prepared per every ten samples within the batch. The maximum relative percent difference between the two results was < 10.00 percent.

3.2 Moisture Measurements

Moisture measurements were taken of all coupons after the initial pressure wash with the 2300 (2100) psi washer and air drying for one week. These measurements were taken to demonstrate that the moisture

level in the coupons was not changing over time and to ensure that all coupons were in a “dry” state before being tested. The measurements were taken for five consecutive days prior to any deposition and indicated that the surfaces of the coupons were dry throughout the measurement period. The moisture of the asphalt coupons could not be measured due to incompatibility with the moisture meter. However, the average moisture measurements for the brick and concrete coupons were 0.13 percent and 1.49 percent, respectively.

3.3 Quality Control Samples

QC samples generated during testing included use of positive control coupons, blank coupons, and high pressure water blank samples. Blank coupons were coupons that had not been dosed with any contaminant. The high pressure water blank samples were taken directly from the bottom of the chamber. Positive controls were created by cutting Ziploc® plastic bags into three-inch squares after the bags were washed and cleaned using Triton-X cleaning solution. These squares were placed (overlapping) on the surface of a blank large coupon so that the entire surface of the coupon was covered with the plastic Ziploc® material. There were a total of 25 Ziploc squares for each positive control deposition. The deposition blank coupon was covered by a 5 x 5 array of Ziploc® squares overlapping each other. The actual deposition area for the 3 x 3 array of holes in the template was 36 in². The CsCl contaminant solution was deposited using the high pressure syringe. After depositing the CsCl solution into all deposition locations, all squares were placed into a clean plastic 1000-mL beaker. A plastic beaker was used to insure no absorption of Cs on the silica in the glass beaker. A 1 % HNO₃ solution was used to fill the beaker to approximately 360 mL. Paraffin was placed on top of the beaker to prevent any loss of HNO₃; the beaker was then sonicated for 15 minutes to promote the release of Cs particles into the solution. After sonication, each Ziploc® square was removed from the beaker and discarded. HNO₃ was added to the beaker up to a 450-mL total volume. Using an Eppendorf pipette, 0.5 mL was extracted from the beaker and placed into a clean 50-mL vial. The vial was then filled to a volume of 50 mL with DI water and submitted for analysis by ICP-MS using EPA Standard Method 200.8. A modified EPA Method 200.8 was used for analysis since EPA 200.8 specifications do not list Cs as a measured component.

The average recoveries for the Cs positive controls were between 70 percent and 120 percent. The RSD of Cs amounts from positive control recovery results was less than 19 percent. The background levels from the blank coupons were subtracted from the test coupon results. The average Cs removed for the brick blank coupons was 0.652 µg with a standard deviation of 0.175. The average Cs removed for the concrete blank coupons was 0.466 µg with a standard deviation of 0.225. The average Cs removed for the asphalt blank coupons was 0.061 µg with a standard deviation of 0.047. The rinsate from the brick blank chamber washes was analyzed for Cs, and the average Cs obtained was 0.159 µg with a standard deviation of 0.121. The rinsate from the concrete blank chamber washes was analyzed for Cs and the average Cs obtained was 0.155 µg with a standard deviation of 0.150. The rinsate from the asphalt blank chamber washes was analyzed for Cs, and the average Cs obtained was 0.129 µg with a standard deviation of 0.128.

The ANCOVA model was also used to analyze the blank results. The results indicated orientation was not a significant predictor for either liquid or solid removal. For liquid removal, pressure was significant for brick only. For solid removal, pressure was significant for brick and concrete only. Mean liquid removal was similar in brick and concrete and was greater than for asphalt. Mean solid removal was similar for asphalt and concrete, which were both less than for brick. These results should be used with caution

because brick and concrete blanks were not tested at the lower pressures, and ANCOVA models are predicting negative results for solids at lower pressures.

4 Results

4.1 Cesium Removal Efficacy

Efficacy tables and comparison plots are presented for each material in the following subsections. The results are presented as total removal efficacy. Solid Cs amount is the amount of Cs removed from the coupon via the solid phase of the rinsate. Liquid Cs amount is the amount of Cs removed from the coupon via the liquid phase of the rinsate. The total Cs removal efficacy is the sum of solid Cs efficacy (M_{SR}) and liquid Cs efficacy (M_{LR}) as shown in Section 2.9.

4.1.1 Overview: Brick

Cesium removal efficacy results for brick are listed in Table 4-1. The removal efficacy results reflect the average of three coupons. The solid and liquid Cs amount removed was blank coupon-corrected using a bulk average blank for each material. If the resultant solid/liquid Cs amount removed resulted in a negative value, a “zero” was used for that solid/liquid quantity.

Table 4-1. Test Conditions and Removal Efficacy Results for Brick

Coupon ID	Nozzle Pressure (psi)	Deposition (Wet/Dry)	Coupon Orientation (H/V*)	Total Cs Removal Efficacy (%)
252-03	2100	Dry	H	14.2
255-03	2100	Dry	V	17.2
259-03	4000	Dry	H	30
262-03	4000	Dry	V	27.5
265-03	5000	Dry	H	34.3
268-03	5000	Dry	V	34.9
216-03	6000	Dry	H	30.1
219-03	6000	Dry	V	29.8
222-03	7000	Dry	H	31.8
225-03	7000	Dry	V	27.7
234-03	2100	Wet	V	8.3
231-03	2300	Wet	H	5.1
237-03	4000	Wet	H	14.1
240-03	4000	Wet	V	15.6
243-03	5000	Wet	H	15
246-03	5000	Wet	V	20.9
201-03	6000	Wet	H	19.3
204-03	6000	Wet	V	16.8
207-03	7000	Wet	H	20.2
210-03	7000	Wet	V	23.3

*Horizontal/Vertical

The following figures illustrate the Cs removal efficacy results for the brick pressure washing tests. Figures 4-1 and 4-2 show the results for the wet deposition method and the horizontal and vertical orientations, respectively. Figures 4-3 and 4-4 show the same results for the dry deposition method. In addition, each chart shows the error bars with one standard deviation. Note that each data point for efficacy represents the average for a set of triplicate test results.

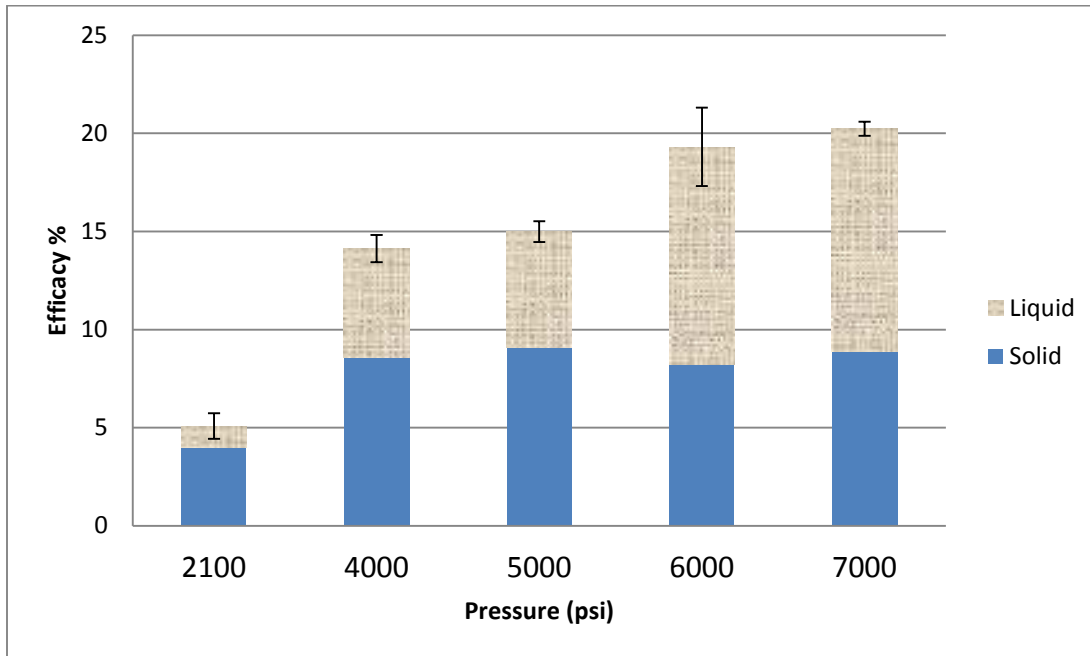


Figure 4-1. Removal efficacy from brick – horizontal orientation and wet deposition

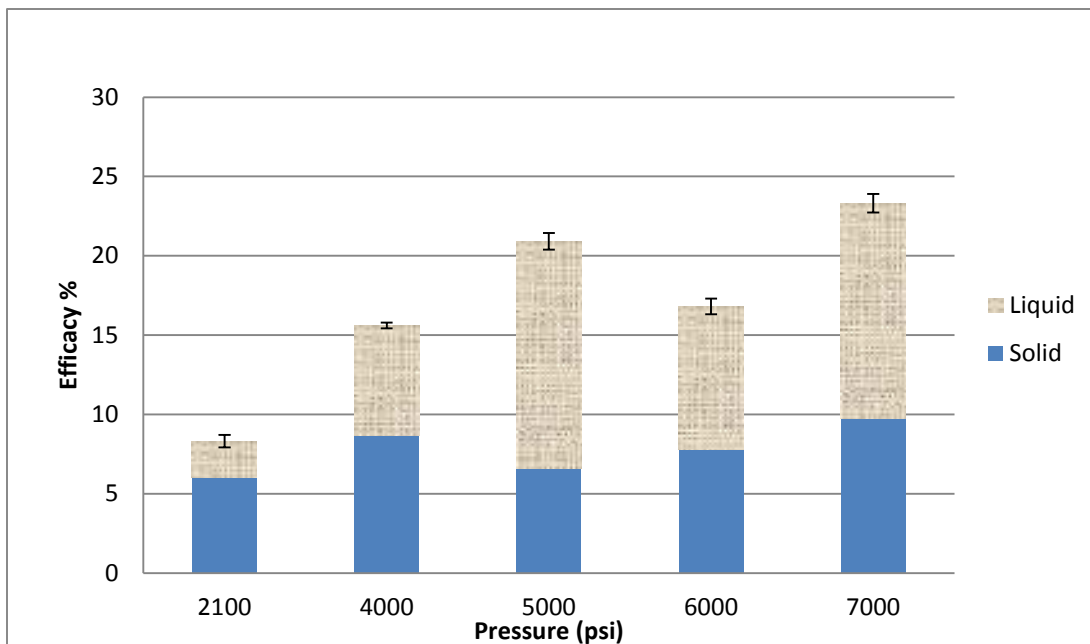


Figure 4-2. Removal efficacy from brick – vertical orientation and wet deposition

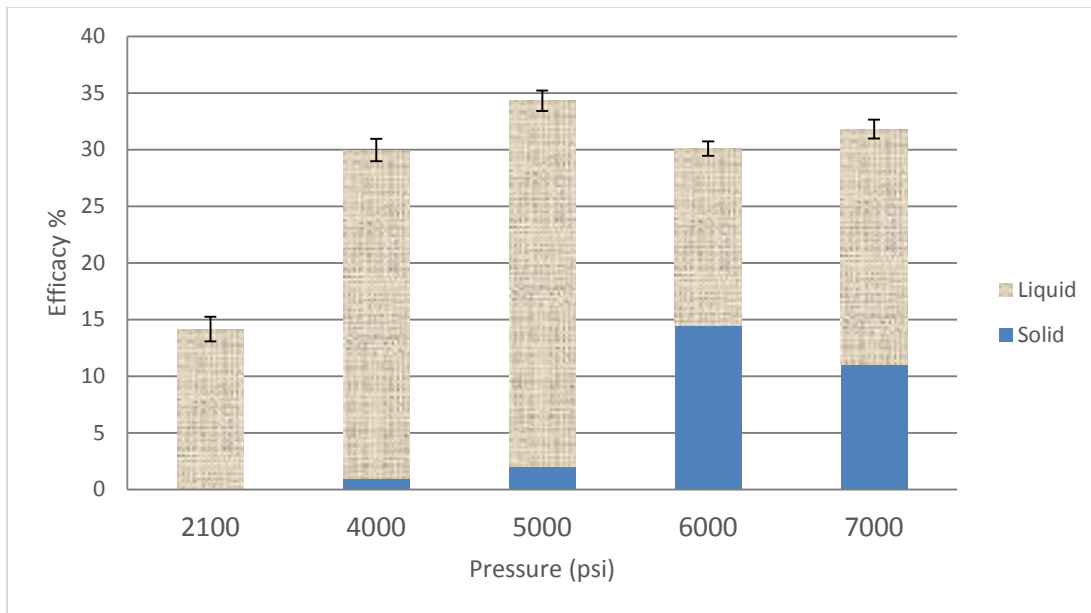


Figure 4-3. Removal efficacy from brick – horizontal orientation and dry deposition

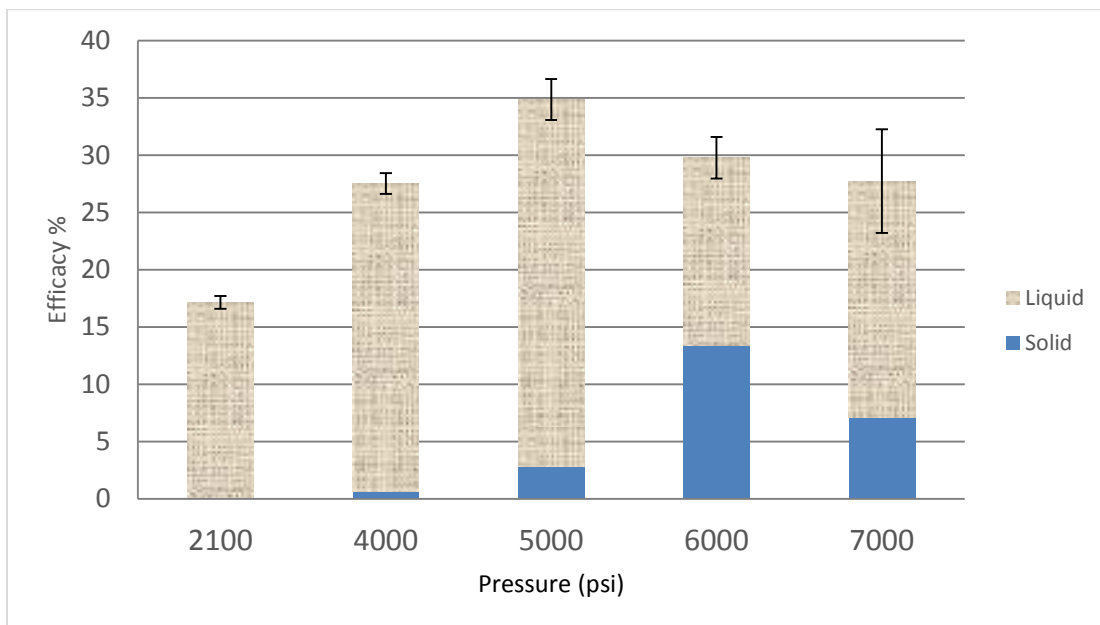


Figure 4-4. Removal efficacy from brick – vertical orientation and dry deposition

4.1.2 Overview: Concrete

Cesium removal efficacy results for concrete are listed in Table 4-2. The removal efficacy results reflect the average of three coupons. The solid and liquid Cs amount removed was blank coupon-corrected using a bulk average blank for each material. If the resultant solid/liquid Cs amount removed resulted in a negative value, a “zero” was used for that solid/liquid quantity.

Table 4-2. Test Conditions and Removal Efficacy Results for Concrete

Coupon ID	Nozzle Pressure (psi)	Deposition (Wet/Dry)	Coupon Orientation (H/V)	Total Cs Removal Efficacy (%)
122-03	2100	Dry	H	15.1
125-03	2100	Dry	V	19
129-03	4000	Dry	H	15.5
132-03	4000	Dry	V	21.5
135-03	5000	Dry	H	24.9
138-03	5000	Dry	V	10.3
160-03	6000	Dry	H	13.9
163-03	6000	Dry	V	17.8
184-03	7000	Dry	H	14.7
187-03	7000	Dry	V	24.2
101-03	2100	Wet	H	12.2
104-03	2100	Wet	V	5.6
107-03	4000	Wet	H	18
110-03	4000	Wet	V	13.4
113-03	5000	Wet	H	13.9
116-03	5000	Wet	V	16.8
144-03	6000	Wet	V	15.2
147-03	6000	Wet	H	16.6
151-03	7000	Wet	H	21.1
154-03	7000	Wet	V	14.3

The following figures illustrate the Cs removal efficacy results for the concrete pressure washing tests. Figures 4-5 and 4-6 show the results for the wet deposition method for the horizontal and vertical orientations, respectively. Figures 4-7 and 4-8 show the same results for the dry deposition method. In addition, each chart shows the error bars with one standard deviation. Note that each data point for efficacy represents the average for a set of triplicate test results.

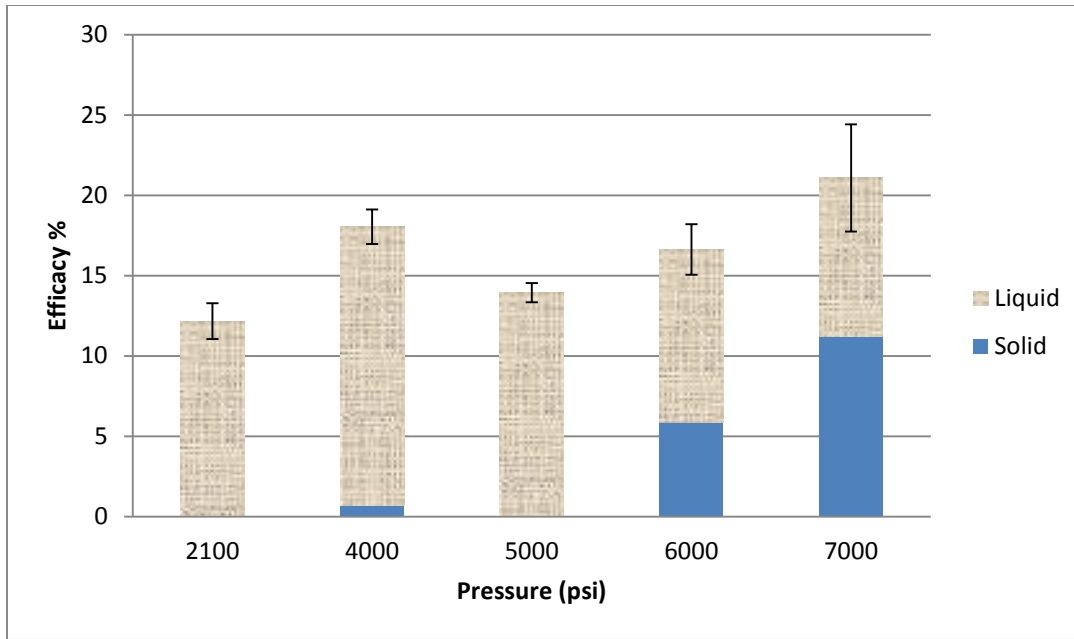


Figure 4-5. Removal efficacy from concrete – horizontal orientation and wet deposition

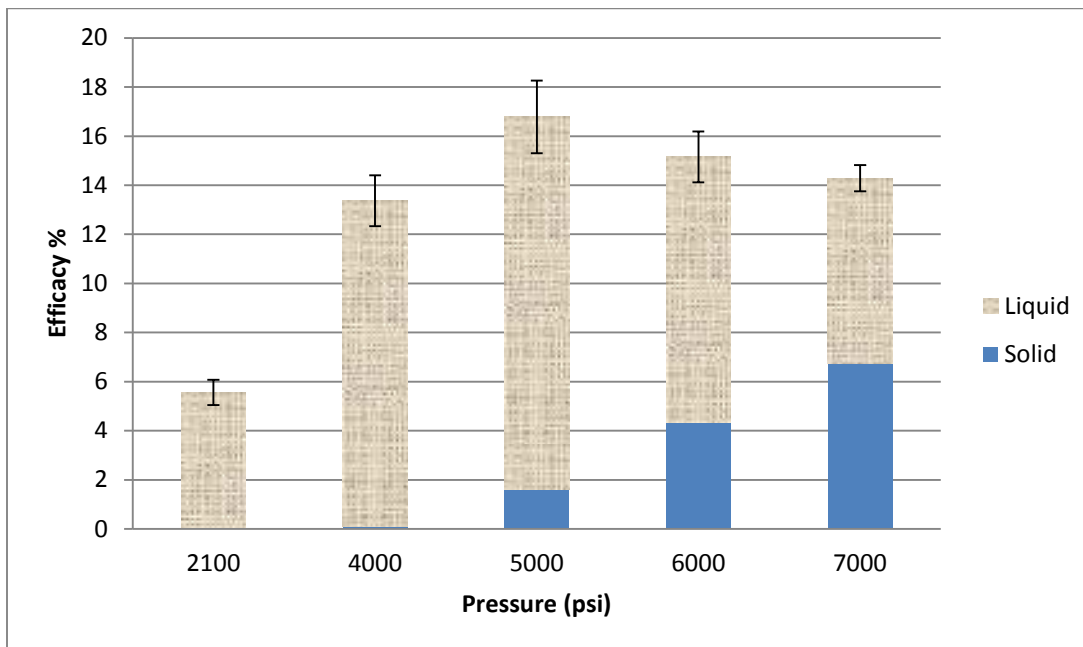


Figure 4-6. Removal efficacy from concrete – vertical orientation and wet deposition

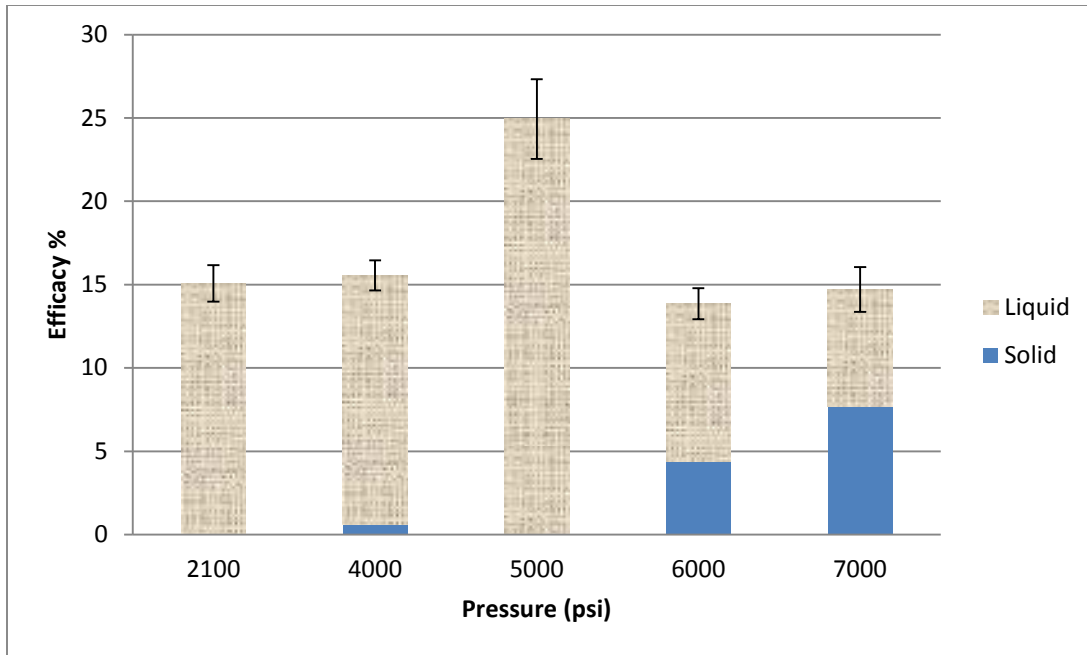


Figure 4-7. Removal efficacy from concrete – horizontal orientation and dry deposition

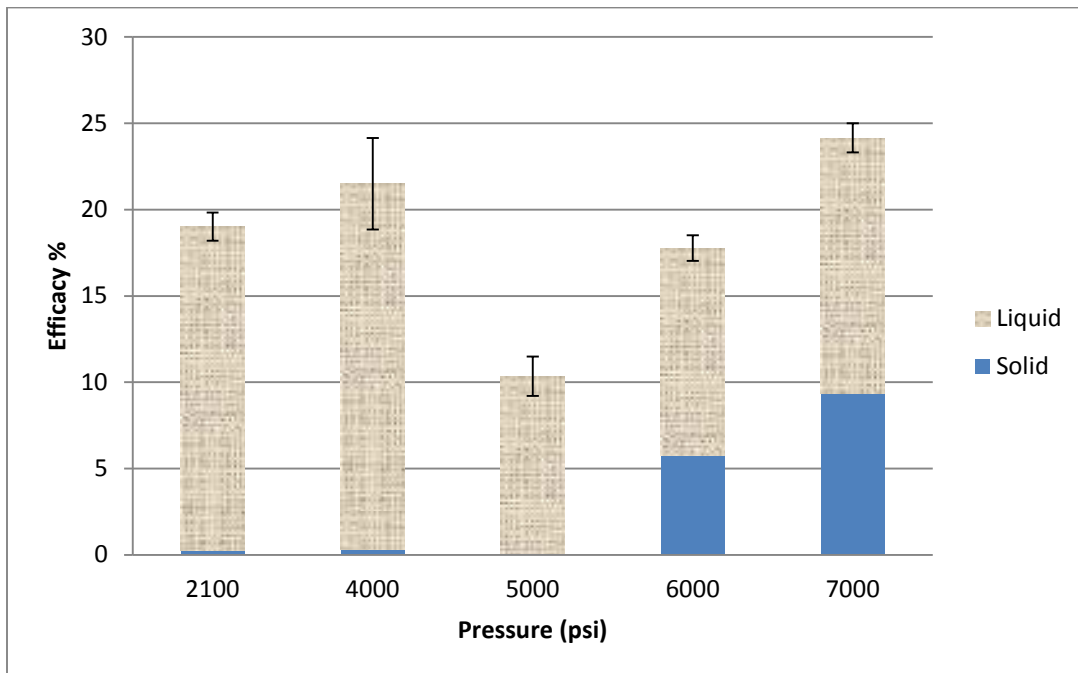


Figure 4-8. Removal efficacy from concrete – vertical orientation and dry deposition

4.1.3 Overview: Asphalt

Cesium removal efficacy results for asphalt are listed in Table 4-3. The removal efficacy results reflect the average of six coupons. The solid and liquid Cs amount removed was blank coupon-corrected using a bulk average blank for each material. If the resultant solid/liquid Cs amount removed resulted in a negative value, a “zero” was used for that solid/liquid quantity.

Table 4-3. Test Conditions and Removal Efficacy Results for Asphalt

Coupon ID	Nozzle Pressure (psi)	Deposition (Wet/Dry)	Total Cs Removal Efficacy (%)
006-03	2100	Dry	45.1
034-03	4000	Dry	51.5
037-03	5000	Dry	57.3
091-03	6000	Dry	73.1
094-03	7000	Dry	78.3
001-03	2100	Wet	45.9
013-03	4000	Wet	55.1
025-03	5000	Wet	42.1
048-03	6000	Wet	55.8
086-03	7000	Wet	73.8

The following figures illustrate the Cs efficacy results for the asphalt pressure washing tests. Figures 4-9 and 4-10 show the results for the wet and dry deposition methods for the horizontal orientations, respectively. In addition, each chart shows the error bars with one standard deviation. Note that each data point for efficacy represents the average for a set of six test results.

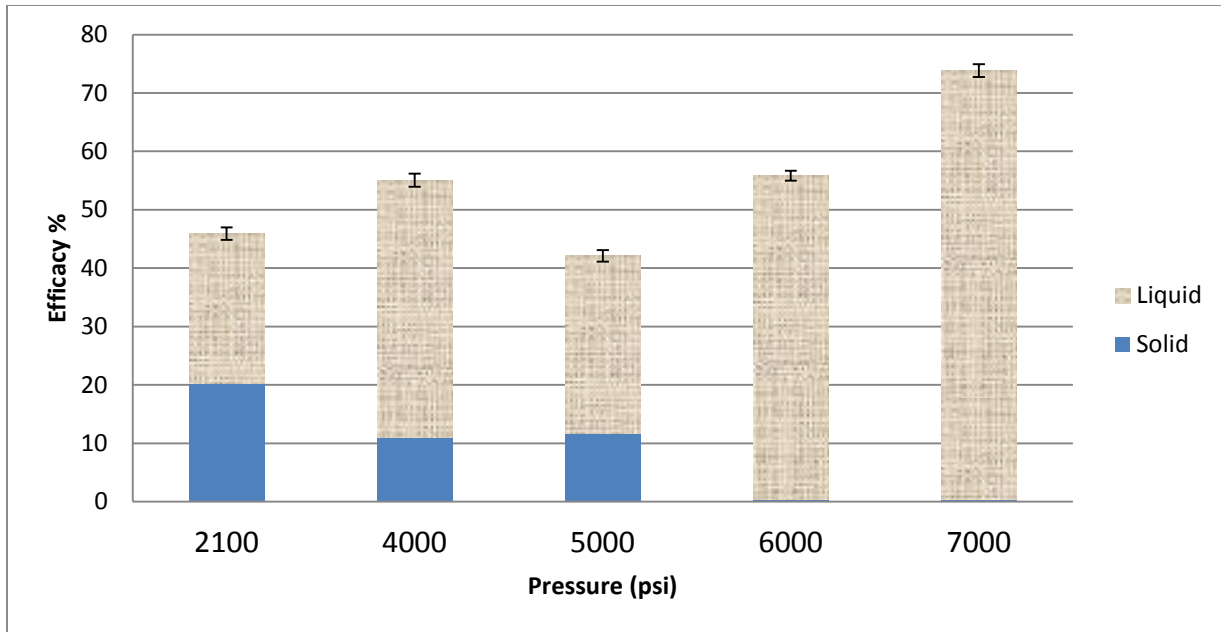


Figure 4-9. Removal efficacy from asphalt – horizontal orientation and wet deposition

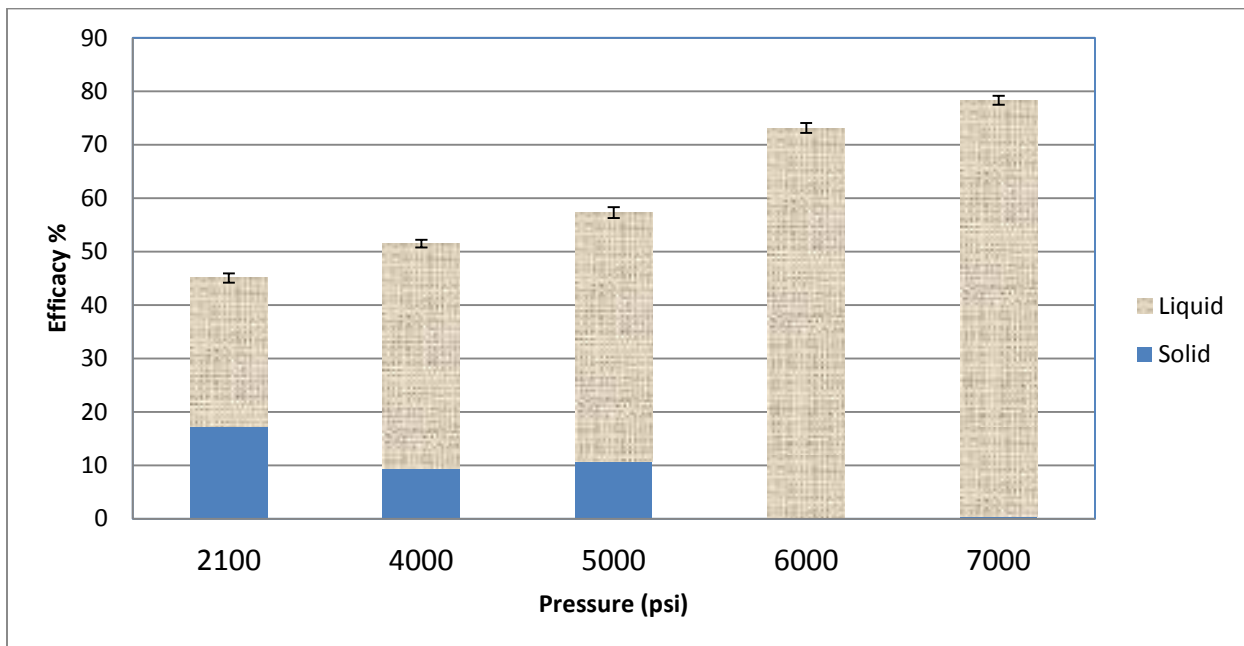


Figure 4-10. Removal efficacy from asphalt – horizontal orientation and dry deposition

4.1.4 Effects of Water Pressure

A total efficacy comparison with a linear regression analysis (Microsoft Excel, Microsoft Corporation, One Microsoft Way, Redmond, WA, 2013) for the Cs removal as a function of water pressure was conducted for the three materials, and the results are shown in Figures 4-11 and 4-12 for all conditions tested. The pressure effect via the ANCOVA model varied by material (but not by deposition method) and was significant only for asphalt. Efficacy increase as a function of water pressure was statistically significant for asphalt ($p = 0.011$) with the dry deposition method. Efficacy as a function of water pressure was not statistically significant for brick ($p = 0.106$) or concrete ($p = 0.969$).

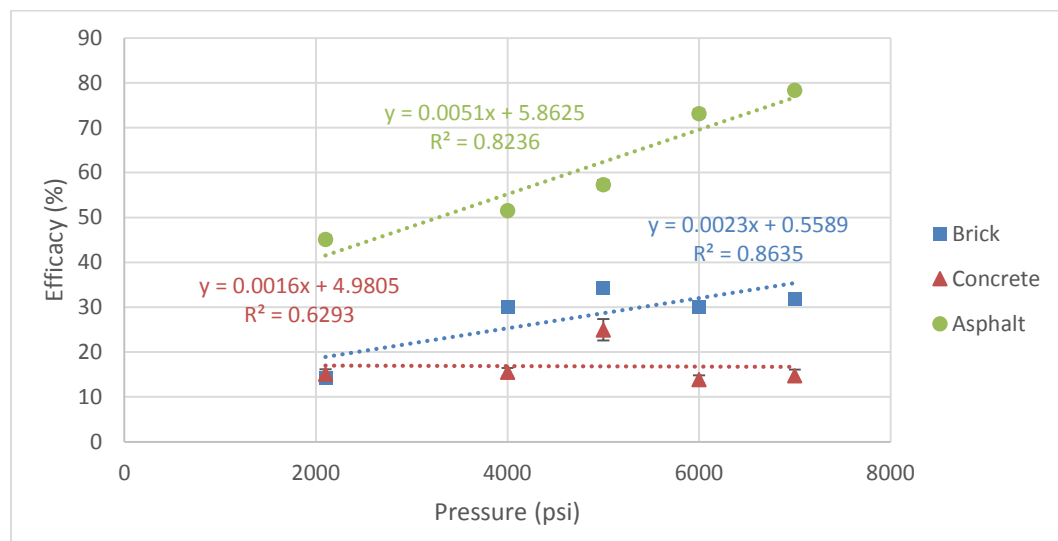


Figure 4-11. Total removal efficacy comparison for horizontal orientation and dry deposition

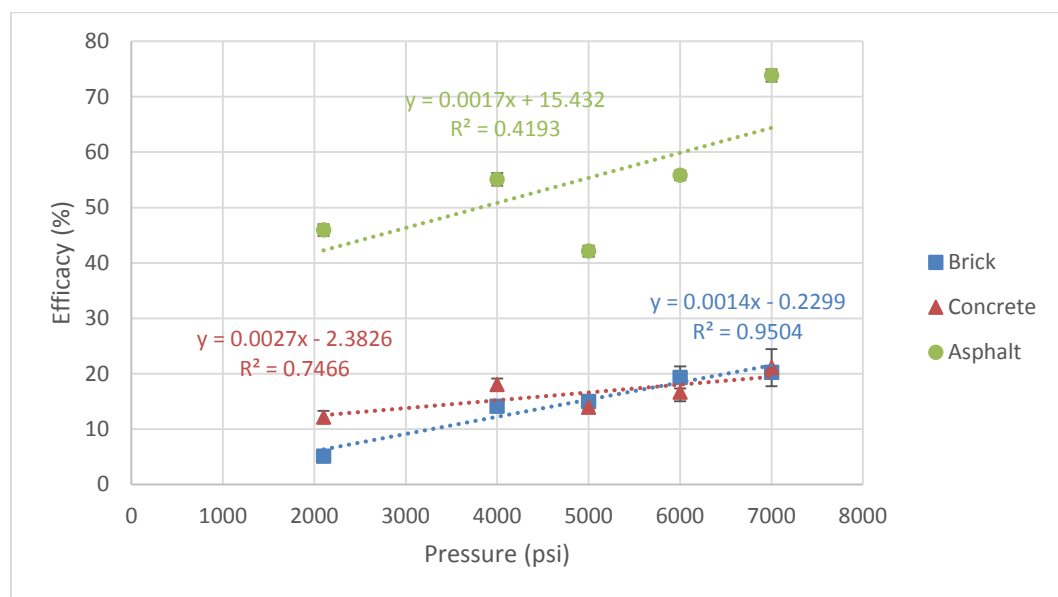


Figure 4-12. Total removal efficacy comparison for horizontal orientation and wet deposition

The total removal efficacy comparison for the horizontal orientation and wet deposition method shows that pressure was extremely significant for brick ($p = 0.005$), but not significant for concrete ($p = 0.130$) nor asphalt ($p = 0.194$). Total removal efficacy ($R^2 = 0.75$) via the ANCOVA model varied by material and deposition and was greatest for asphalt. The pressure effect evaluated by the ANCOVA model varied by material, but not by deposition method and was significant only for asphalt and brick.

A comparison of solid Cs efficacy for all three substrates and all conditions tested is shown in Figures 4-13 and 4-14. As shown, the solid efficacy for asphalt decreased with increasing pressure. However, solid efficacy for brick and concrete increased with increasing pressure, most likely due to asphalt being comprised of tar and not smaller particles like brick and concrete. Solid removal efficacy via the ANCOVA model ($R^2 = 0.35$) varied by material but not deposition method and was greatest in asphalt and brick.

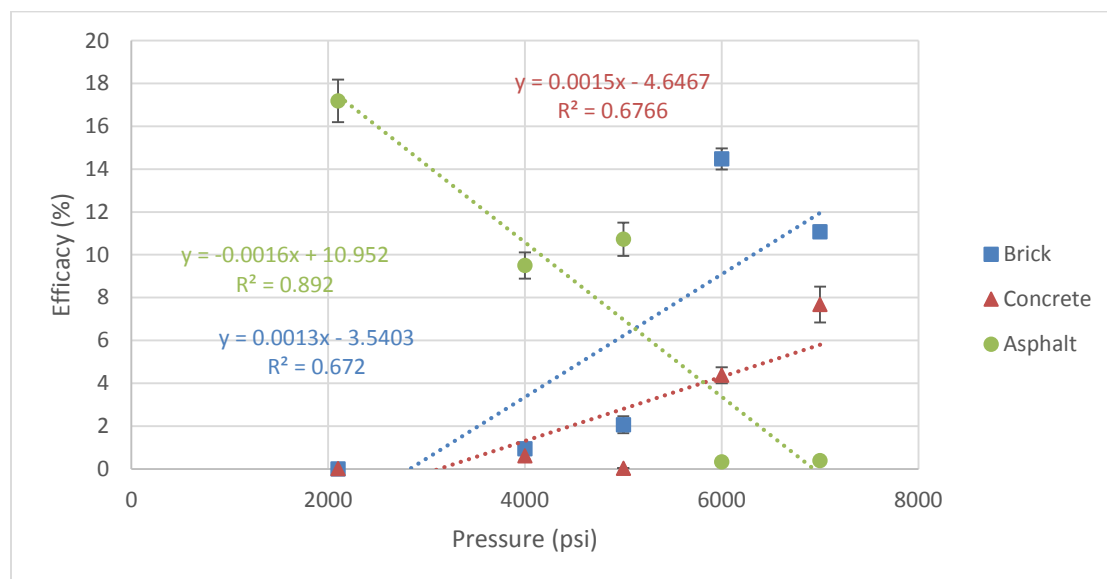


Figure 4-13. Solid efficacy comparison for horizontal orientation and dry deposition

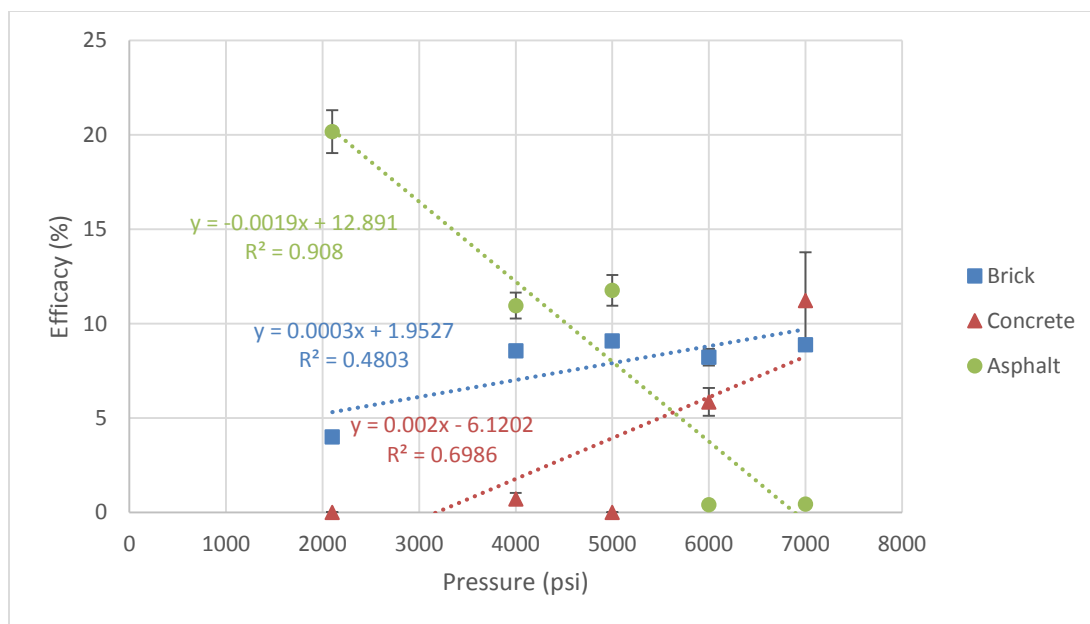


Figure 4-14. Solid efficacy comparison for horizontal orientation and wet deposition

A comparison of the liquid efficacy for all three substrates and all conditions tested is shown in Figures 4-15 and 4-16. The liquid efficacy increased marginally for brick, but not concrete. However, liquid efficacy increased substantially for asphalt with increasing pressure, as opposed to the solid efficacy. For the liquid ANCOVA model ($R^2 = 0.71$), the main effects (material, pressure, and deposition method) were significant. Liquid removal efficacy was greatest in asphalt and this may be due to fewer debris generated compared to brick and concrete coupons.

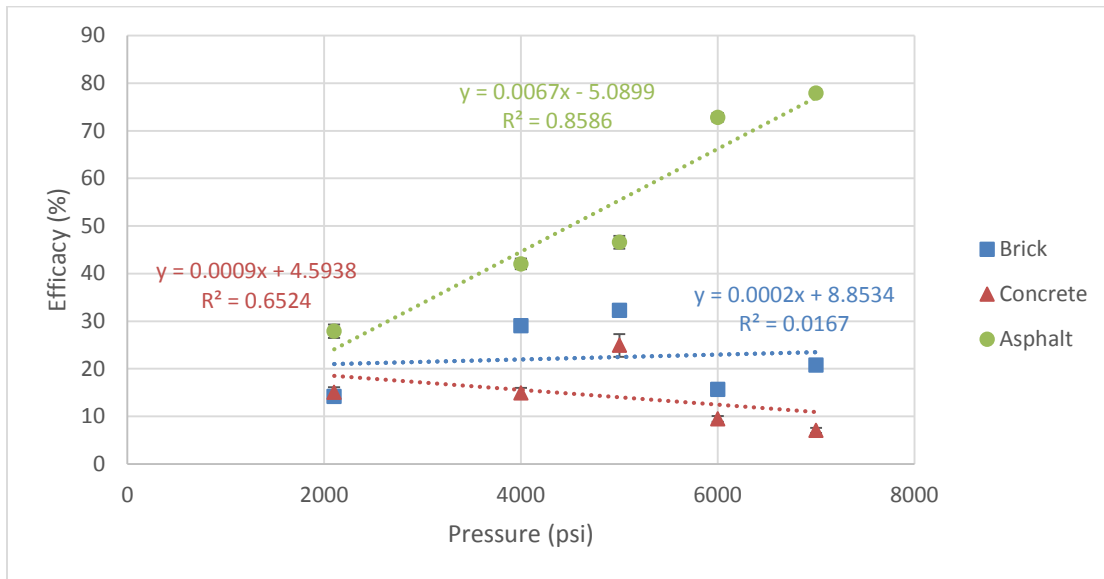


Figure 4-15. Liquid efficacy comparison for horizontal orientation and dry deposition

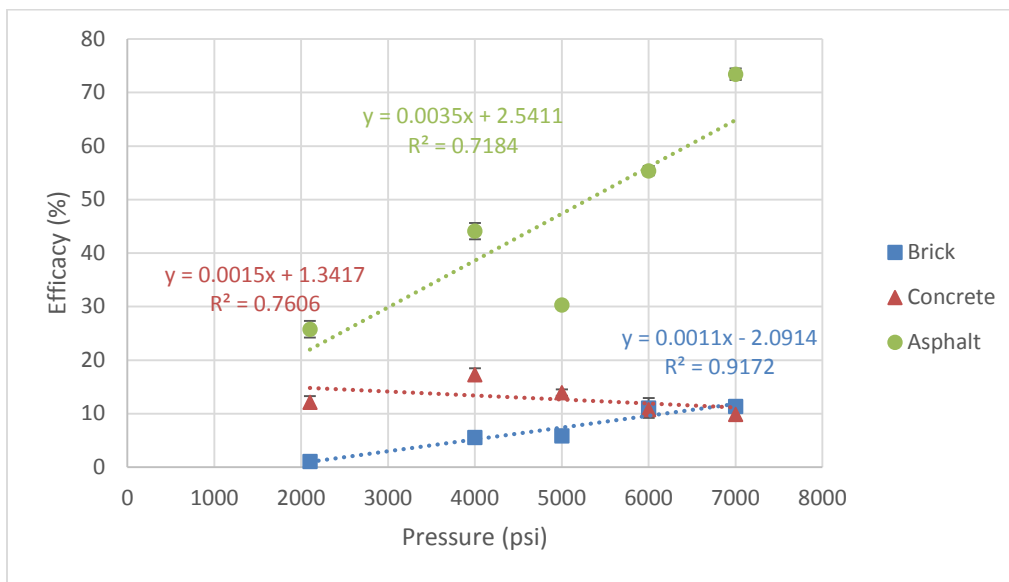


Figure 4-16. Liquid efficacy comparison for horizontal orientation and wet deposition

A total efficacy comparison for the vertical orientation and wet and dry deposition is shown in Figure 4-17. For wet deposition, brick showed increasing efficacy with increasing pressure, and concrete showed decreasing efficacy at 6000 and 7000 psi. For dry deposition, the brick efficacy decreased at 6000 and 7000 psi. The concrete efficacy for the dry deposition decreased at 5000 psi, but increased at 6000 and 7000 psi. For the vertical orientation and wet deposition method, pressure was significant for brick ($p = 0.039$) but not significant for concrete ($p = 0.113$). However, pressure was not significant for brick ($p =$

0.215) nor concrete ($p = 0.802$) in the vertical orientation with the dry deposition method. Results from the ANCOVA model indicated orientation was not a significant factor for either liquid or solid removal.

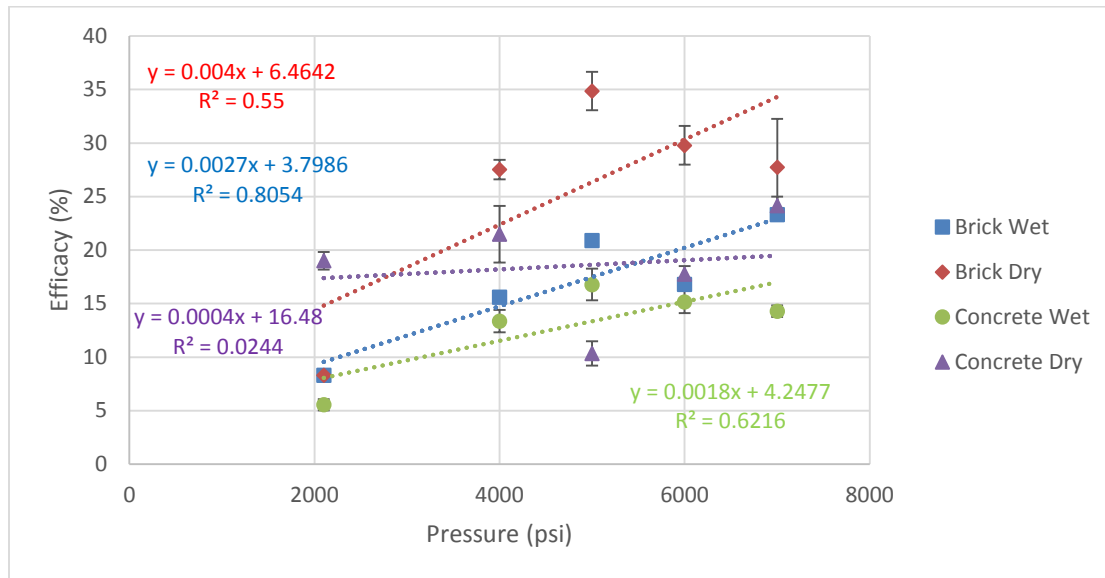


Figure 4-17. Total efficacy comparison for vertical orientation and wet/dry deposition

5 Summary and Recommendations

This study investigated the impact of water pressure conditions for decontamination of urban surfaces contaminated with Cs. The study was conducted to evaluate the decontamination efficacy of pressure washer as functions of water pressure, surface type, surface orientation, and deposition method. The evaluation was performed by contaminating common urban surfaces with nonradioactive CsCl particles and then washing them in a chamber simulating the delivery of high pressure water. The amount of Cs in the liquid and solid portions of the water rinsate samples were measured and analyzed for the impact of individual wash conditions.

The current study found the following key information for surface Cs removal using a pressure washer:

- Increased pressure improved Cs removal efficacy for all three surfaces. Asphalt showed a much more profound effect with increasing pressure than brick and concrete.
- The Cs removal mechanism was further investigated by measuring removal efficacy in solid and liquid portion separately. The results showed that when comparing solid efficacy for all three substrates for the same conditions, asphalt showed decreasing contribution of solid efficacy with increasing pressure, but brick and concrete showed increasing contribution of solid efficacy with increasing pressure. The cause of negative correlation for asphalt's solid efficacy with increased water pressure needs further investigation. Liquid efficacy comparison for the same conditions showed the same profound effect for asphalt as total efficacy, whereas brick and concrete showed minimal increase in efficacy with increasing pressure.
- Total removal efficacy varied by surface types and deposition methods. Asphalt showed the highest Cs removal efficacy (42 % to 78 %), followed by brick (5 % to 35 %) and concrete (6 % to 25 %). Higher Cs removal was observed from dry (methanol) deposition than from wet (water) deposition for all three surfaces, possibly related to the penetration depth of Cs after deposition. Methanol evaporates faster than water, and as a result, the majority of Cs remains close to the surface and is easier to remove.
- Orientation was not a significant factor for either liquid or solid removal for all three surfaces.

The dry and wet deposition test results demonstrated that delayed application of the pressure washer may lead to less removal of Cs from the surface due to the subsurface penetration which may increase over time. For asphalt, Cs removal in the liquid phase was dominant, and the removal efficacy was increased with increased water pressure up to 7000 psi. The water pressure (6000 to 7000 psi) will provide high decontamination efficacy (50 % to 80 %) with minimal destruction. In the case of brick and concrete, test results showed minimal increase (less than 10 % increase) in Cs removal as a function of pressure in the range of 4000 to 7000 psi. However, increased removal efficacy for brick and concrete were related mainly to removal of solids from the coupon by the high pressure stream. Extensive layer removal from brick and concrete surfaces is expected to increase the removal efficacy due to the subsurface penetration and Cs sorption on the removed surfaces. For effective concrete and brick surface decontamination using pressure washer, it will be necessary to assess two items: Cs subsurface distribution and surface removal thickness as a function of water pressure.

6 References

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